Intensities of surface spin wave excitations in inelastic electron scattering

J. Rajeswari,[1,2,*] E. Michel,[1,2] H. Ibach,[2,3] and C. M. Schneider[1,2]
[1]*Peter Grünberg Institut (PGI-6), Forschungszentrum Jülich, 52425 Jülich, Germany*
[2]*Jülich Aachen Research Alliance, 52425 Jülich, Germany*
[3]*Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany*
(Received 14 August 2013; revised manuscript received 17 February 2014; published 28 February 2014)

The intensity of surface spin wave excitations in inelastic electron scattering is measured as function of electron energy for fcc and hexagonal close packed (hcp) cobalt layers. Intensities are converted into scattering probabilities with the help of a recently established calibration of our spectrometer. The scattering probability as function of energy exhibits a peak around 7 and 3 eV for fcc and hcp cobalt, respectively, and decays to immeasurably small values at energies above 10 to 15 eV in stark contrast to theoretical predictions. By comparison to phonon scattering the peaks in the scattering probabilities at low energies are tentatively attributed to image potential induced resonances.

DOI: 10.1103/PhysRevB.89.075438 PACS number(s): 75.30.Ds

I. INTRODUCTION

Inelastic electron scattering or electron energy loss spectroscopy (EELS) has been used for several decades to explore diverse elementary excitations at surfaces such as surface phonons [1–3] and surface plasmons [4,5]. The possibility of using EELS for the studies of surface spin waves was predicted by Mills as early as 1967 [6]. A later theoretical calculation indicated that the probability for inelastic scattering of electrons from spin waves is about three orders of magnitude smaller than that from vibration modes. Nevertheless, it was concluded that spin waves should be observable in loss spectra [7,8]. Motivated by these predictions, advanced electron spectrometers were specifically designed to investigate surface spin wave excitations in the high wave vector regime [9,10]. Over the last decade, the spectrometers have been successfully brought to bear in studies of surface spin waves on ultrathin cobalt and iron films [11–20].

So far the interest focused on the physical properties of surface spin waves as such. This paper is devoted to the observed intensities in inelastic electron scattering. Specifically, we have studied the intensities of surface spin waves as function of energy for epitaxial fcc and hexagonal close packed (hcp) cobalt layers. The experimental data are converted into scattering probabilities $dP/d\Omega$ and compared to theory inasmuch as it exists. The conclusion is a twofold one and rather unexpected. First, at higher electron energies (>30 eV) theory overestimates the scattering probability by at least an order of magnitude. Second, existing surface spin wave data are based on an intensity-enhancement phenomenon at low electron energies whose nature is not understood. By comparison to phonon scattering one might speculate that the enhancement may be due to image potential induced resonances.

II. EXPERIMENTAL

Our experiments were performed on two structurally different cobalt surfaces—fcc and hcp cobalt obtained by growing Co on Cu(100) and Cu(111) substrates, respectively [21,22]. The copper templates are prepared by repeated cycles of sputtering with 1.5 keV Ar atoms followed by annealing to about 1000 K. Cobalt was deposited using electron beam stimulated evaporation from high purity rods onto the Cu substrates held at room temperature. Well-ordered fcc and hcp structures are observed in low energy electron diffraction (LEED). In both cases, no measurable shift in the LEED spot positions occurred upon deposition of cobalt indicating pseudomorphic growth. The thicknesses of the deposited layers are calibrated by the observation of well defined oscillations in the intensity of diffracted beams of 3 keV electrons on the Cu(100) substrate. It is known that Co/Cu(111) exhibits pyramidal island growth after first-layer nucleation rather than layer-by-layer growth [22]. Hence the growth of Co on Cu(111) does not show any intensity oscillations [23]. We therefore used the intensity oscillations from Co/Cu(100) to calibrate the evaporator flux. The thickness calibration for the hcp layers on Cu(111) was obtained by scaling the thickness vs flux calibration to the 15.5% higher layer density on Cu(111). After deposition the samples were transferred into the chamber housing the electron spectrometer. Small amounts of hydrogen and molecular CO are flashed off by annealing briefly to 450 K. After that treatment no traces of contamination were visible in vibration spectroscopy using EELS which is an extremely sensitive test for all common impurities.

The energy loss experiments were carried out with our high resolution electron energy loss spectrometer [10]. The spectrometer is calibrated in terms of the accepted solid angle and the transmission probability as described in Ref. [24]. This enables the conversion of experimental data on intensities into probabilities for each electron to be inelastically scattered by a spin wave.

III. RESULTS

Figure 1 represents two sample spectra measured on a 6 monolayer (ML) Co/Cu(111) surface for a particular in-plane wave vector transfer of $\Delta K_{\parallel} = 0.6$ Å^{-1}. The wave vector transfer is obtained by rotating the sample manipulator while keeping the scattering angle fixed at 90°. The spectrum taken at $E_0 = 3$ eV electron impact energy shows the intense signature of a spin wave peaking at 105 meV (open squares in Fig. 1).

*r.jayaraman@fz-juelich.de

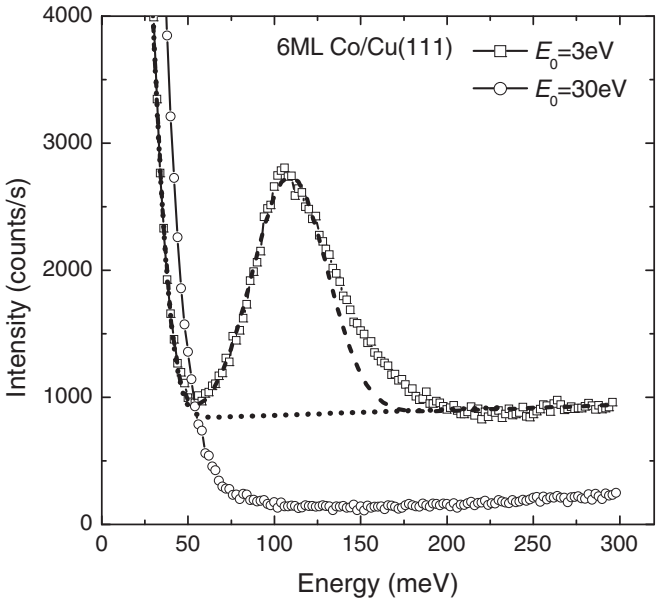

FIG. 1. Electron energy loss spectra measured for 6 ML Co deposited on Cu(111) for an in-plane wave vector transfer $\Delta K_{\parallel} = 0.6$ Å^{-1} along the $\overline{\Gamma}\,\overline{\mathrm{K}}$ direction at electron impact energies of 3 (squares) and 30 eV (circles).

No indication of a spin wave is visible in the spectrum taken at 30 eV for the same wave vector transfer (open circles in Fig. 1). For the purpose of our quantitative study of the spin wave intensities, spectra are fitted to a Gaussian (dashed line in Fig. 1) after subtracting a background (dotted line in Fig. 1). As seen from Fig. 1, fitting to a Gaussian neglects the asymmetry of the spin wave loss which is due to a contribution of standing modes of the film [18,25]. However the fitting procedure suffices for the present purpose.

In Fig. 2(a), we show the energy dependence of spin wave intensities for electrons scattered inelastically from the 8 ML Co/Cu(100) surface. The spin wave intensities are obtained for a wave vector transfer of $\Delta K_{\parallel} = 0.69$ Å^{-1} directed along two high symmetry directions $\overline{\Gamma}\,\overline{\mathrm{X}}$ and $\overline{\Gamma}\,\overline{\mathrm{M}}$. The inset shows the surface Brillouin zone (SBZ) of the fcc(100) surface with the high symmetry directions indicated. The spectral intensities exhibit a peak around 6 eV for the $\overline{\Gamma}\,\overline{\mathrm{X}}$ direction with a rapid decay on either side of this energy and a double-peak structure for the $\overline{\Gamma}\,\overline{\mathrm{M}}$ direction. No discernible spin wave features were found for energies above 15 eV. The error bars in Fig. 2(a) mark the upper limit for a spin wave signal, which is conservatively estimated as the total count rate at the spin wave energy minus the minimum in the background.

A similar peaking of the intensity at lower energy is observed for the hcp cobalt film. Figure 3(a) shows the energy dependence of the surface spin wave signal for 6 ML Co/Cu(111) for a wave vector of $\Delta K_{\parallel} = 0.55$ Å^{-1} directed along the $\overline{\Gamma}\,\overline{\mathrm{K}}$ direction of the SBZ. The peak in the intensity now occurs around 3 eV. Again, no discernible spin wave signals are observed for higher impact energies. The intensities in Figs. 2(a) and 3(a) are converted into the energy integrated

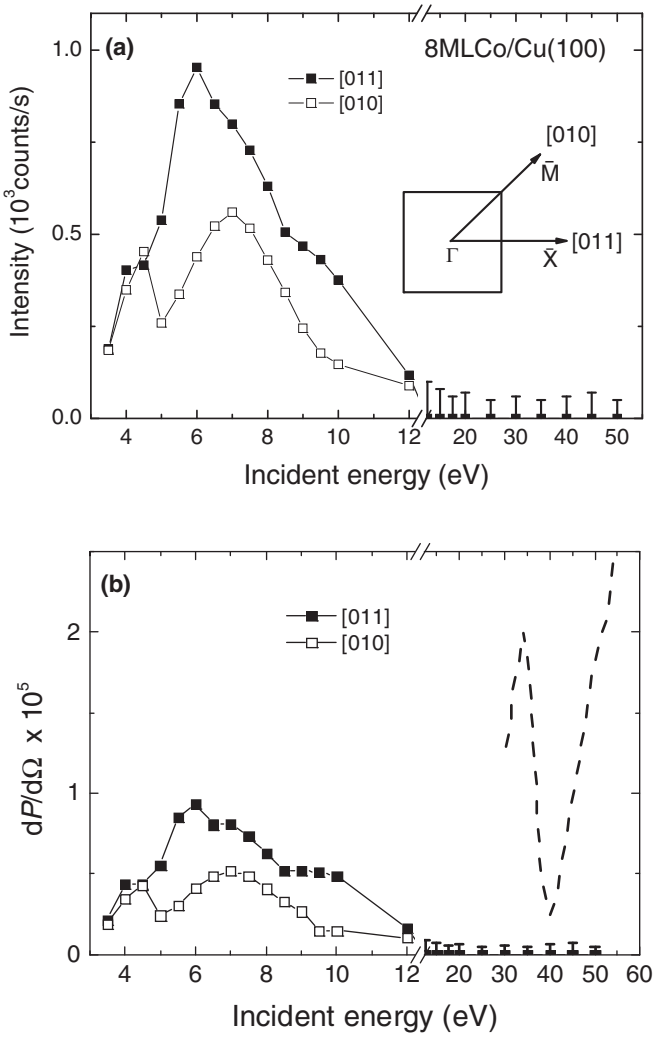

FIG. 2. (a) Energy dependence of the spin wave peak intensity of an 8 ML Co film deposited on a Cu(100) surface for $\Delta K_{\parallel} = 0.69$ Å^{-1} measured along the $\overline{\Gamma}\,\overline{\mathrm{X}}$ and $\overline{\Gamma}\,\overline{\mathrm{M}}$ directions [surface Brillouin zone is shown as an insert in (a)]. No discernible spin wave features are observed above 15 eV. The error bars mark the upper limit estimated from the noise. (b) Spectral intensities converted into scattering probability per solid angle for inelastic scattering. The dashed line shows the spin wave intensities calculated for a wave vector transfer $\Delta K_{\parallel} = 0.66$ Å^{-1} for the Fe(100) surfaces according to Gokhale *et al.* [7].

scattering probability per solid angle defined as [7]

$$\frac{dP}{d\Omega} = \int_{-\infty}^{\infty} \frac{d^2P}{d\hbar\omega d\Omega} d\hbar\omega. \tag{1}$$

It was shown in [24] that $dP/d\Omega$ is calculated from the experimental intensities by

$$\frac{dP}{d\Omega} = \frac{I_{\mathrm{peak}} s_{\omega}}{I_{\mathrm{in}} T \Delta\Omega s_{\mathrm{el}}}, \tag{2}$$

in which I_{peak} is the peak count rate of the spin wave peak at $\hbar\omega$, and I_{in} is the number of electrons impinging on the surface per time. The ratio of variances s_{ω} and s_{el} characterizing the Gaussian widths of the loss peak and the elastic diffuse peak, respectively, are determined from the spectra. The quantity

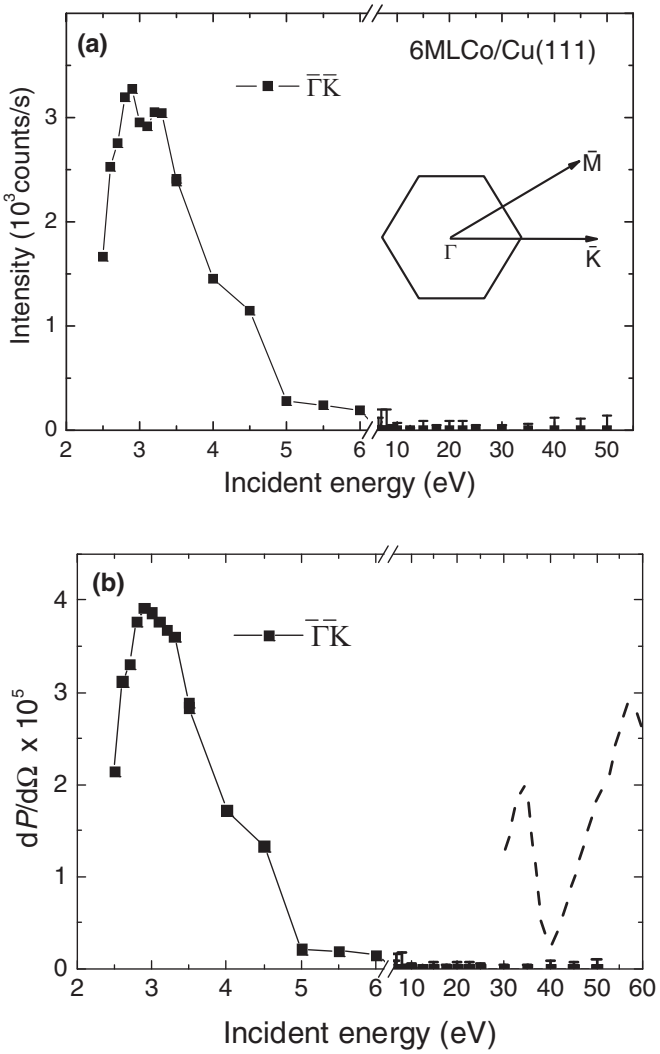

FIG. 3. (a) Energy dependence of the spin wave peak intensity of a 6 ML Co film deposited on a Cu(111) surface for $\Delta K_{\parallel} = 0.55$ Å^{-1} ($\Delta K_{\parallel} = 0.6$ Å^{-1} for $E_0 > 10$ eV) measured along the $\overline{\Gamma}\,\overline{\mathrm{K}}$ direction [surface Brillouin zone is shown as an insert in (a)]. No discernible spin wave features are observed above 10 eV. The error bars mark the upper limit estimated from the noise. (b) Spectral intensities converted into scattering probability per solid angle for inelastic scattering. The dashed line shows the spin wave intensities calculated for a wave vector transfer $\Delta K_{\parallel} = 0.66$ Å^{-1} for the Fe(100) surfaces according to Gokhale *et al.* [7].

$T\,\Delta\Omega$ is the product of the transmission probability and the solid angle probed by the spectrometer, respectively. We have calculated this quantity as function of electron energy with the help of extensive electron optical calculations which include the diffuse scattering from the sample. For our spectrometer $T\,\Delta\Omega$ is described by the parameterized equation {see Fig. 16 in combination with Eq. (6) of [24]}

$$10^4 T_{\mathrm{L}}\Delta\Omega = 38.9 + 10.8\exp(-0.05E_0/\mathrm{eV}). \qquad (3)$$

Using the above equations, the intensities are converted into the scattering probability per solid angle $dP/d\Omega$ and the result is depicted in Figs. 2(b) and 3(b). Because of the smooth calibration function (3), $dP/d\Omega$ exhibits the same trend as the intensities. For comparison we have plotted the scattering probability per solid angle calculated by Gokhale *et al.* [7] for the Fe(100) surfaces and a wave vector transfer $\Delta K_{\parallel} = 0.66$ Å^{-1}.

IV. DISCUSSION

The low scattering probability which we have found for energies above 10 to 15 eV is at variance with the theory for spin wave scattering of Gokhale *et al.* [7]. These authors have calculated the scattering probability for Fe(100) surfaces in the energy range of 30 to 150 eV. According to that work the scattering probability displays oscillations characteristic of inelastic electron diffraction. The mean value of the scattering probability is about 2×10^{-5}, hence more than an order of magnitude higher than the upper bound calculated from our experimental data in the higher energy range. Since cobalt and iron have roughly the same magnetic moments the discrepancy cannot be attributed to the difference in the material but must rather lie in a fundamental defect of the theory. One reason for the failure of the theory could be the use of the Heisenberg model which is known to be inadequate for itinerant magnets [26,27].

The peak of the intensity around 6 eV for the fcc surface along the $\overline{\Gamma}\,\overline{\mathrm{X}}$ direction was already observed by Vollmer *et al.* in 2004 [28]. At the time, the peak was considered to possibly arise from an increase in exchange scattering probability at lower energies [29,30] in combination with the falling sensitivity of the spectrometer at lower energies. Our calibration, however, shows that at least for our spectrometer the sensitivity does not decrease for lower energies [Eq. (3)]. The double peak structure of the intensity vs energy for the $\overline{\Gamma}\,\overline{\mathrm{M}}$ direction as well as the observation of similar peaking of the intensity, albeit at a lower energy on hcp cobalt, points towards a fundamentally different, yet general nature of the phenomenon. In search for an explanation we note that the cross section for electron scattering from phonons displays similar peak structures at low energies. The cross section as function of beam energy for the symmetric stretching mode of hydrogen atoms adsorbed on W(001) at saturation coverage, e.g., exhibits a pronounced peak around 4.7 eV and a multitude of very sharp fine structures above 4.7 eV. These structures were attributed to image potential induced resonances [31,32]. The peak and the fine structure arise from a temporary trapping of incident or scattered electrons in the image potential in a region within a few angtroms above the surface [32,33]. Due to that temporary trapping, the interaction time of the electron with the surface is longer than in normal electron-surface scattering resulting in an overall increase in the scattering probability. In addition, fine structures may arise from the oscillating phase match of the incident and scattered electron. By analogy to those studies we suggest that the peak and fine structures in Figs. 2 and 3 may be likewise due to image potential induced resonances.

V. CONCLUSION

For energies higher than 10 to 15 eV the experimentally observed scattering probability for spin waves is at

least an order of magnitude lower than predicted by existing theory. At low electron energies, the scattering probability shows a resonance behavior, which by analogy to similar resonances in phonon scattering may be attributed to image potential resonances. However, a quantitative theory of the phenomenon applicable to spin waves is lacking. This is a rather unsatisfactory situation in view of the fact that all experimental data on surface spin waves are obtained with impact energies in the resonance range.

ACKNOWLEDGMENT

J.R. gratefully acknowledges the financial support from NRW Research School "Forschung mit Synchrotronstrahlung in den Nano- und Biowissenschaften."

[1] H. Ibach and D. L. Mills, *Electron Energy Loss Spectroscopy and Surface Vibrations* (Academic Press, New York, 1982).
[2] S. Lehwald, J. M. Szeftel, H. Ibach, T. S. Rahman, and D. L. Mills, Phys. Rev. Lett. **50**, 518 (1983).
[3] M. Balden, S. Lehwald, H. Ibach, A. Ormeci, and D. L. Mills, J. Electron Spectrosc. Relat. Phenom. **64–65**, 739 (1993).
[4] K.-D. Tsuei, E. Plummer, A. Liebsch, E. Pehlke, K. Kempa, and P. Bakshi, Surf. Sci. **247**, 302 (1991).
[5] K. Pohl, B. Diaconescu, G. Vercelli, L. Vattuone, V. M. Silkin, E. V. Chulkov, P. M. Echenique, and M. Rocca, Europhys. Lett. **90**, 57006 (2010).
[6] D. L. Mills, J. Phys. Chem. Solids **28**, 2245 (1967).
[7] M. P. Gokhale, A. Ormeci, and D. L. Mills, Phys. Rev. B **46**, 8978 (1992).
[8] M. Plihal and D. L. Mills, Phys. Rev. B **58**, 14407 (1998).
[9] H. Ibach, D. Bruchmann, R. Vollmer, M. Etzkorn, P. S. Anil Kumar, and J. Kirschner, Rev. Sci. Instrum. **74**, 4089 (2003).
[10] H. Ibach, J. Rajeswari, and C. M. Schneider, Rev. Sci. Instrum. **82**, 123904 (2011).
[11] R. Vollmer, M. Etzkorn, P. S. Anil Kumar, H. Ibach, and J. Kirschner, Phys. Rev. Lett. **91**, 147201 (2003).
[12] R. Vollmer, M. Etzkorn, P. S. Anil Kumar, H. Ibach, and J. Kirschner, J. Magn. Magn. Mater. **272–276**, 2126 (2004).
[13] M. Etzkorn, P. S. Anil Kumar, R. Vollmer, H. Ibach, and J. Kirschner, Surf. Sci. **566**, 241 (2004).
[14] W. X. Tang, Y. Zhang, I. Tudosa, J. Prokop, M. Etzkorn, and J. Kirschner, Phys. Rev. Lett. **99**, 087202 (2007).
[15] C. L. Gao, A. Ernst, G. Fischer, W. Hergert, P. Bruno, W. Wulfhekel, and J. Kirschner, Phys. Rev. Lett. **101**, 167201 (2008).
[16] J. Prokop, W. X. Tang, Y. Zhang, I. Tudosa, T. R. F. Peixoto, K. Zakeri, and J. Kirschner, Phys. Rev. Lett. **102**, 177206 (2009).
[17] K. Zakeri, Y. Zhang, J. Prokop, T.-H. Chuang, N. Sakr, W. X. Tang, and J. Kirschner, Phys. Rev. Lett. **104**, 137203 (2010).
[18] J. Rajeswari, H. Ibach, C. M. Schneider, A. T. Costa, D. L. R. Santos, and D. L. Mills, Phys. Rev. B **86**, 165436 (2012).
[19] J. Rajeswari, H. Ibach, and C. M. Schneider, Phys. Rev. B **87**, 235415 (2013).
[20] J. Rajeswari, H. Ibach, and C. M. Schneider, Europhys. Lett. **101**, 17003 (2013).
[21] C. M. Schneider, P. Bressler, P. Schuster, J. Kirschner, J. J. de Miguel, and R. Miranda, Phys. Rev. Lett. **64**, 1059 (1990).
[22] K. Heinz, S. Müller, and L. Hammer, J. Phys.: Condens. Matter **11**, 9437 (1999).
[23] Z. Q. Qiu, J. Pearson, and S. D. Bader, Phys. Rev. B **46**, 8195 (1992).
[24] H. Ibach and J. Rajeswari, J. Electron Spectrosc. Relat. Phenom. **185**, 61 (2012).
[25] J. Rajeswari, H. Ibach, and C. M. Schneider, Phys. Rev. Lett. (to be published).
[26] A. T. Costa, R. B. Muniz, and D. L. Mills, Phys. Rev. B **70**, 054406 (2004).
[27] A. T. Costa, R. B. Muniz, and D. L. Mills, Phys. Rev. B **69**, 064413 (2004).
[28] R. Vollmer, M. Etzkorn, P. S. Anil Kumar, H. Ibach, and J. Kirschner, Thin Solid Films **464–465**, 42 (2004).
[29] J. Glazer and E. Tosatti, Solid State Commun. **52**, 905 (1984).
[30] H. Hopster, R. Raue, and R. Clauberg, Phys. Rev. Lett. **53**, 695 (1984).
[31] R. F. Willis, Surf. Sci. **89**, 457 (1979).
[32] B. M. Hall, S. Y. Tong, and D. L. Mills, Phys. Rev. Lett. **50**, 1277 (1983).
[33] E. G. McRae, Rev. Mod. Phys. **51**, 541 (1979).